

# Probing Scattering Phases via Two-Center Interferences in Collisions of $\text{He}^{2+}$ on CO

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Phases of quantum transition amplitudes contain very important information about physical processes. However, as a rule, they may not be accessed experimentally. This is, in particular, the case when collisions of ions and atoms are studied. In this letter we explore double-electron capture in collisions of alpha particles with CO molecules. Regarding the atomic cores of the molecule as two "slits", we show that atomic "double-slit" interference can be exploited in order to experimentally extract information about phases of the projectile scattering on the cores.

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In ion-atom collision processes, interactions between particles are often manifested in terms of phase factors in the transition amplitude. These phases contain important information about the collision process but are in many cases still poorly understood. Unfortunately, they cannot be directly accessed in experiments on ion-atom collisions since cross sections – the quantities measurable in experiment – are proportional to the absolute square of the transition amplitude.

In this paper, we report on an alternative method with which the relative scattering phase of ion-atom collisions can be read out via two-center interference effects.

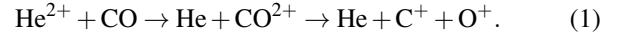
Effects of two-center interference in ion-molecule collisions was first predicted in capture collisions between the proton and  $\text{H}_2$  molecule where the mechanical double slit is replaced by a diatomic molecule [1]. The prediction was confirmed in electron capture by  $\text{O}^{8+}$  from the homonuclear diatomic molecules (*homo*-DM)  $\text{H}_2$  [2] and later also in the electron energy spectra in single ionization of  $\text{H}_2$  induced by the impact of 60 MeV/u  $\text{Kr}^{34+}$  [3]. In the latter case, the molecular orientation was not experimentally determined. However, even under such conditions the authors of [3] found signatures of interferences by considering the ratio between the experimental results and corresponding cross sections for independent hydrogen atoms.

With the advent of reaction microscopes, the determination of the complete kinematic information in ion-atom/molecule collisions became feasible [4, 5]. As a result, much more pronounced two-center interference fringes in cross sections for dissociative capture [6] and excitation [7] were observed in  $\text{H}_2^+ + \text{He}$  collisions. Currently there exists extensive literature on two-center interference effects occurring in collisions of molecules with ions (e.g. [8–14]), electrons (e.g. [15–18]) and photons (e.g. [19–21]).

However, so far most studies on ion-molecule collisions focused merely on the role of the geometric-kinematic interference phase factor  $\exp(i\mathbf{q} \cdot \mathbf{R})$ , where  $\mathbf{q}$  is the momentum transfer and  $\mathbf{R}$  is the internuclear vector of a diatomic molecule. Only in [6] and [7], an additional  $\pi$  phase-shift was explored

and attributed to different symmetries of the initial and final electronic states of the molecule.

In this work, we explore double electron capture in collisions of alpha particles with the hetero-diatomic molecule (*hetero*-DM) CO,



By regarding the atomic cores of the molecule as two "slits", we show that atomic "double-slit" interference in collisions with molecules can be exploited in order to experimentally extract information about phases of the projectile scattering on the cores of the atoms constituting the molecule.

Atomic units are used throughout unless otherwise is stated.

The experiment was performed using the reaction microscope of the Institute of Modern Physics, Lanzhou [22]. Briefly,  $\text{He}^{2+}$  ions produced in the electron cyclotron resonance ion source were extracted, charge analyzed, and then accelerated to energies of 30 and 135 keV/u, respectively. The well-collimated ion beam entered into the collision chamber and was vertically crossed with a neutral carbon monoxide beam from a supersonic gas jet. After the collision, the projectiles were charge-state analyzed by an electrostatic deflector downstream the collision center. The (undeflected) neutralized projectiles were detected by a position sensitive detector, while the charged projectile beam components were directed away from the detector. The primary beam was collected by a Faraday cup. The charged molecule fragments produced in the collision region were extracted by an electric field of 120 V/cm perpendicular to both the projectile and the jet directions. All fragments with energies up to 12 eV were guided to a position sensitive detector mounted at the end of a time-of-flight (TOF) spectrometer. Three dimensional momenta of the fragments can thus be calculated with the recorded position and timing information. To achieve high accuracy in the molecular orientation,  $\text{C}^+$  and  $\text{O}^+$  were detected simultaneously although detecting just one fragment would be sufficient to determine the molecular orientation.

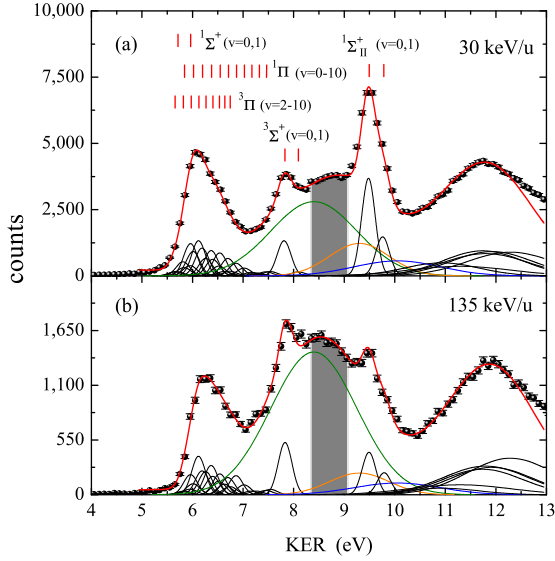


FIG. 1. (Color online) KER of fragments resulting from  $\text{CO}^{2+}$ . The positions of the vibrational resolved red lines are from [24]. Fitting curves are possible contributions from different states of  $\text{CO}^{2+}$  (for details see text).

Another important requirement to determine the molecular orientation with sufficient accuracy is that the  $\text{CO}^{2+}$  molecular ion dissociates much faster than it rotates, e.g. the so-called axial recoil approximation (ARA) [23] must be valid. It should be noted that in spite of the target cooling one cannot assume that the molecule is always in the rotational ground state for two reasons: a) in the plane perpendicular to the jet expansion the gas is only cooled by geometric collimation, which only cools the gas translationally, but not rotationally. b) Angular momentum can be transferred to the molecule in the collision itself. Out of many dissociative  $\text{CO}^{2+}$  states, the ARA can be applied to just a few repulsive states which are free of any potential wells. In the following, we shall denote these states as the fast repulsive states and the states involving potential wells as the slow dissociating states.

During the Coulomb explosion of the  $\text{CO}^{2+}$  molecule potential energy is converted to the kinetic energy of the fragments (kinetic energy release, KER). Each electronic state of  $\text{CO}^{2+}$  corresponds to a characteristic KER-distribution. Figure 1 shows the KER spectra obtained for collisions at impact energies of 30 keV/u and 135 keV/u. For both collision energies, the KER spectra possess certain qualitative similarities. For instances, each of the spectra consists of three narrow peaks centered approximately at 6.1, 7.8 and 9.4 eV. Besides, a ‘shoulder’-like structure around 8.5 eV and a broad peak centered at about 11.8 eV are also present for both impact energies.

In the KER spectra, the peak centered at 7.8 eV is solely from the vibrational ground state levels of  $^3\Sigma^+$  [24]. By performing Gaussian fittings to the peak, we estimate the resolution of the KER to be about 0.24 eV full width at half

maximum (FWHM). This value was further used for fitting the KER distributions of other vibrational dissociative states. For instance, the slow dissociating states of  $^1\Sigma^+$  ( $v=0,1$ ),  $^1\Pi$  ( $v=1-10$ ) and  $^3\Pi$  ( $v=2-10$ ) contribute to the peak at KER=6.1 eV, whereas the peak centered at KER=9.4 eV originates mainly from  $^1\Sigma^+$  ( $v=0,1$ ) states [24].

According to the theoretical calculations [25], the shoulder between 8.3-9.2 eV and the broad maximum centered at 11.8 eV originate from two groups of fast repulsive states ( $^1\Delta$ ,  $^3\Pi_{II}$ ,  $^3\Sigma^-$  etc., and  $^3\Pi_{III}$ ,  $^1\Sigma^-$ ,  $^3\Sigma^-$ ,  $^1\Delta_{II}$  etc.), respectively. The fitting result indicates that the events with  $8.4 \text{ eV} < \text{KER} < 9.1 \text{ eV}$  (the shaded area in Figure 1) are predominantly from the  $^1\Delta$  repulsive state, while the  $^3\Sigma^-$  and  $^3\Pi_{II}$  repulsive states only give merely small contributions. In this regime, the axial recoil approximation is valid and the angular resolution in our experiment is estimated to be about  $10^\circ$  FWHM.

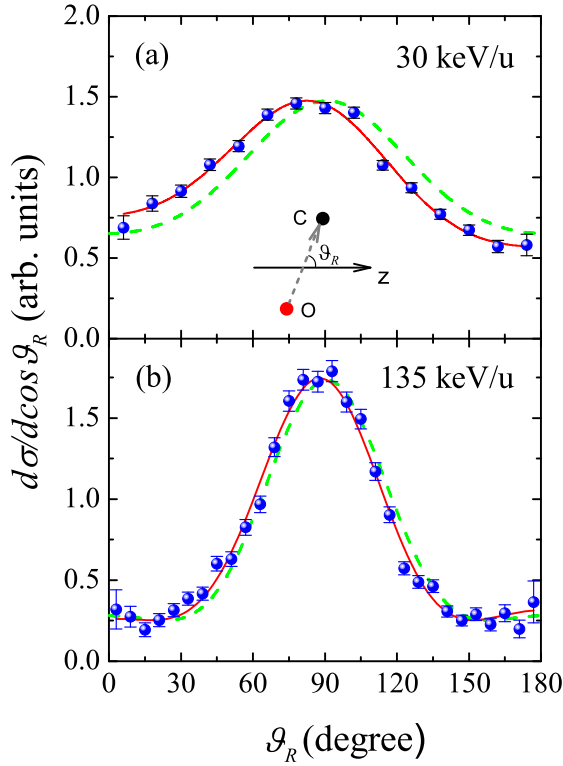
We define our systems of coordinates as follows: the inter-nuclear separation vector  $\mathbf{R}$  of the molecule is pointing from the oxygen to the carbon nucleus, and the molecular orientation is expressed in terms of the relative angle  $\vartheta_R$  between  $\mathbf{R}$  and the direction of the motion of the incident projectile (see inset of Figure ). For instance, with this definition the  $\text{C}^+$  fragment is considered as moving in the forward direction provided  $\vartheta_R < 90^\circ$  and in the backward direction if  $\vartheta_R > 90^\circ$ .

Figure shows the molecular orientation distributions ( $d\sigma/d\cos\vartheta_R = d\sigma/d\Omega$ ) for the marked regime of Figure 1, i.e. from the first group of fast repulsive states (here we refer to  $^1\Delta$ ,  $^3\Pi_{II}$ ,  $^3\Sigma^-$ , etc as the first electronic state group, and discussions for the second group of  $^3\Pi_{III}$ ,  $^1\Sigma^-$ ,  $^3\Sigma^-$ ,  $^1\Delta_{II}$  etc. will be presented in the supplementary material). In the absence of any orientation effects these spectra should be constant. However, in both spectra maxima approximately at  $\vartheta_R = 90^\circ$  are observed, which become increasingly pronounced with increasing projectile energy.

Such maxima observed in collisions between ions and *homo*-DM targets were interpreted as due to two-center interference effects (e.g. [9, 26]). A qualitative difference between our data for the *hetero*-DM CO with earlier data for *homo*-DM is that the present spectra show significant asymmetries in angular distributions about  $\vartheta_R = 90^\circ$ , whereas for the *homo*-DM targets the distributions are symmetric. Furthermore, the asymmetry gradually vanishes as the collision energy increases.

We start our discussion of the basic physics lying behind the experimental results with two remarks. First, the collisions, which are studied, are characterized by small momentum transfers (typically not exceeding several atomic units) such that the recoil velocities of the nuclei of the molecule in the collision are negligible. Second, the collisions are very fast on the molecular time scales.

Therefore, we may assume that the collision occurs at ‘frozen’ positions of the molecular nuclei and for its description we use a reference frame where the (center of mass of the) molecule is initially at rest (=the laboratory frame). If we choose, as the origin, the middle point between the nuclei of the molecule then the coordinates of the nuclei are  $\pm\mathbf{R}/2$



Molecular orientation ( $\vartheta_R$ ) dependent cross sections, i.e.  $d\sigma/d\cos\vartheta_R$  for the collision energies of 30 and 135 keV/u from top to bottom. Red solid lines are fitting results with Eq. (6) and dashed curves are simulations with the same equation but neglect the contribution of  $\Delta'$ .

( $\mathbf{R}$  is the internuclear vector of the molecule).

In order to get insight into the origin of the asymmetries seen in the spectra of Figure , we propose a simple model for the capture process in question. We suppose that there are two pathways for the reaction: in one of them capture of two electrons from the molecule is accompanied by projectile scattering on (interaction with) the atomic core of the center 1 of the molecule whereas in the other the same electrons are captured by the projectile which scatters on the atomic core of center 2.

Within this model the amplitude  $A_{fi}(\mathbf{q})$  for the process can be approximated by [14]

$$A_{fi}(\mathbf{q}) = A_{fi}^{(1)}(\mathbf{q}) \exp\left(-i\frac{\mathbf{q} \cdot \mathbf{R}}{2}\right) + A_{fi}^{(2)}(\mathbf{q}) \exp\left(i\frac{\mathbf{q} \cdot \mathbf{R}}{2}\right), \quad (2)$$

where  $\mathbf{q}$  is the momentum transfer in the collision. In the above expression the partial amplitudes  $A_{fi}^{(1)}(\mathbf{q})$  and  $A_{fi}^{(2)}(\mathbf{q})$  refer to the two pathways of the reaction described above and the terms  $\exp(\pm i\mathbf{q} \cdot \mathbf{R}/2)$  are the geometrical-kinematical factors arising due to the difference in the positions of the molec-

ular centers and reflecting also the momentum transferred in the collision.

The partial amplitudes  $A_{fi}^{(1)}$  and  $A_{fi}^{(2)}$  can be expressed as

$$\begin{aligned} A_{fi}^{(1)} &= |A_{fi}^{(1)}| \exp(i\alpha_1) \\ A_{fi}^{(2)} &= |A_{fi}^{(2)}| \exp(i\alpha_2), \end{aligned} \quad (3)$$

where  $|A_{fi}^{(j)}|$  and  $\alpha_j$  ( $j = 1, 2$ ) are the absolute values and the phases, respectively, of the amplitudes.

One can see that  $\alpha_j$  ( $j = 1, 2$ ) includes all possible phases presented in interactions between the projectile and the corresponding target atom, for instance, the phase induced in the *adiabatic* processes (although very small in fast collisions, but not zero), the Coulomb phase of the ionic core in presence of the fast-moving projectile field, so on and so forth.

Taking into account Eqs.(2)-(3) the (fully differential) cross section for electron capture in collisions with two-atomic molecules can be expressed as

$$\begin{aligned} \sigma_{fi} &= |A_{fi}(\mathbf{q})|^2 \\ &= |A_{fi}^{(1)}(\mathbf{q})|^2 + |A_{fi}^{(2)}(\mathbf{q})|^2 \\ &\quad + 2|A_{fi}^{(1)}(\mathbf{q})||A_{fi}^{(2)}(\mathbf{q})|\cos(\Phi), \end{aligned} \quad (4)$$

where

$$\begin{aligned} \Phi &= \Delta + \mathbf{q} \cdot \mathbf{R} \\ &= \Delta + \mathbf{q}_\perp \cdot \mathbf{R}_\perp + q_\parallel R \cos\vartheta_R. \end{aligned} \quad (5)$$

In the above expressions  $\mathbf{q}_\perp$  and  $q_\parallel$  are the components of the momentum transfer  $\mathbf{q}$  perpendicular and parallel, respectively, to the collision velocity. In double capture process  $q_\parallel = Q/v + v$  where  $Q$  is the binding energy changes of system. Further,  $\Delta = \alpha_2 - \alpha_1$ ,  $\mathbf{R}_\perp$  is the part of the internuclear vector  $\mathbf{R}$  which is perpendicular to the collision velocity and  $\vartheta_R$  is the angle between  $\mathbf{R}$  and the collision velocity.

It is seen in Eqs.(4)-(5) that the cross section is the sum of three terms. Two of them describe the separate contributions to the process due to the two different pathways of the reaction whereas the last term arises due to the interference of these pathways. This term is proportional to  $\cos\Phi$  where the phase  $\Phi$ , in addition to the term  $\mathbf{q} \cdot \mathbf{R}$ , depends also on  $\Delta = \alpha_2 - \alpha_1$ . The appearance of the extra term is caused by the differences in the interaction between the projectile and the atomic cores 1 and 2 of the molecule.

In collisions with homo-nuclear molecules, if odd numbers of electrons are captured from (or excited to) the *ungerade* state, a  $\pi$  phase shift will also enter into  $\Delta$  due to the parity conservation [6, 7]. Apparently, such phase due to the symmetries of the molecular states has no projectile velocity dependence. In our case, these phases, if exist, can also enter into  $\Delta$ . However, the measured phase has a clear projectile velocity dependence which rules out such possibilities.

We note that when one considers collisions with single atoms the phases of the transition amplitudes drop out from

the cross section. Therefore, although such phases contain important information about collision processes, they cannot be accessed in experiments on ion-atom collisions.

However, as we see, the situation may drastically change if collisions with molecules are explored instead of/in addition to collisions with atoms. Therefore, studying collisions with hetero-nuclear molecules offers also a perspective of an additional insight into the dynamics of collisions with atoms by determining the (relative) phases experimentally.

Taking into account the above discussion we fit our experimental results using the following expression

$$\frac{d\sigma_{ji}}{d\cos\vartheta_R} = \sigma (1 + \lambda \cos(\psi \cos\vartheta_R + \Delta')). \quad (6)$$

Here,  $\sigma$  is regarded as the incoherent sum of the contributions to the process from the separated atoms,  $\lambda$  is a parameter which determines the visibility of the interference between the two pathways,  $\psi = q_{\parallel}R$ , and the phase shift  $\Delta'$  is understood as the difference between the scattering phases averaged over the transverse momentum transfer.

For each of the impact energies, the phase shift  $\Delta'$  can be obtained by fitting the corresponding experimental data with Eq.(6) using  $\Delta'$  and  $\psi$  as free parameters. The best fits, shown by solid curves in Figure , are obtained by setting  $\psi = 2.39 \pm 0.17$  and  $3.42 \pm 0.06$ , and  $\Delta' = -0.31 \pm 0.04$  and  $-0.13 \pm 0.03$  for collision energies of 30 and 135 keV/u, respectively. In each spectrum we also plot the cross sections obtained by setting  $\Delta' = 0$  in Eq.(6) while keeping  $\psi$  unchanged. The corresponding results are shown by dashed curves.

Thus, the asymmetry observed in the experimental data can be understood as a distortion effect on two-center interference. The decrease of the asymmetry with increasing the impact energy can be explained by noting that in ion-atom collisions at intermediate impact energies the effective strength of the interaction between the projectile and the atomic cores is expected to be roughly inversely proportional to the projectile velocity  $v$ , accordingly we obtain  $\Delta' \propto 1/v$ .

From our results it follows that  $\Delta'_{30\text{keV/u}}/\Delta'_{135\text{keV/u}} \approx 2.4 \pm 0.6$  which agrees reasonably well with the velocity ratio  $v_{135\text{keV/u}}/v_{30\text{keV/u}} \approx 2.1$  within the experimental resolution. We thus can conclude that the asymmetry in the experimental spectra is caused by the difference in the effective strength of the interactions between the projectile and the atomic cores that leads to an additional phase shift. Furthermore, with increasing collision velocity the asymmetry eventually disappears because of the weakening of the effective strength of the interactions.

In two-center interference experiment the effective internuclear distance  $\mathbf{R}$  can not be simply taken as the equilibrium internuclear distance [9], the fitting values of  $\psi$  thus can not be directly verified in our work. However, such difficulty can be removed by comparing the ratio of  $\psi_{30\text{keV/u}}/\psi_{135\text{keV/u}}$  which gives  $q_{\parallel,30\text{keV/u}}R/[q_{\parallel,135\text{keV/u}}R] = q_{\parallel,30\text{keV/u}}/q_{\parallel,135\text{keV/u}}$ . The theoretical value gives  $q_{\parallel,30\text{keV/u}}/q_{\parallel,135\text{keV/u}} \approx 0.75$  and the

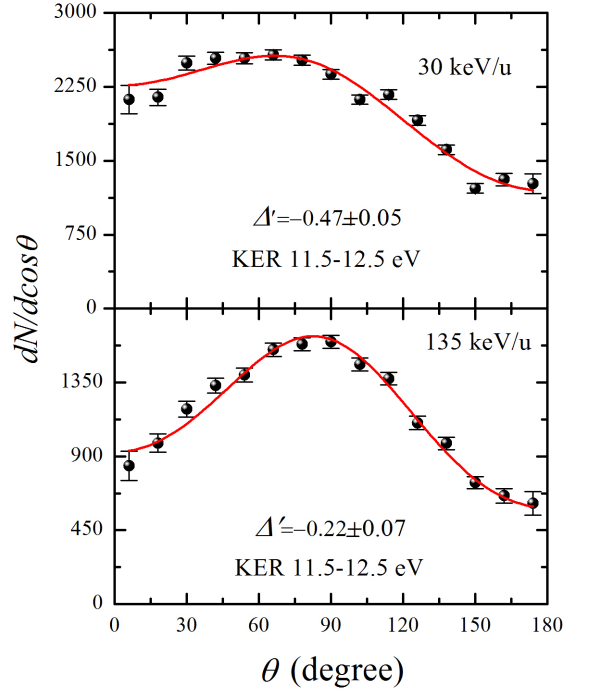


FIG. 2. (Color online) Molecular orientation ( $\theta_R$ ) dependent cross sections of  $11.5 \text{ eV} < \text{KER} < 12.5 \text{ eV}$  for the collision energies of 30 and 135 keV/u from top to bottom. Red solid lines are fitting results with Eq. 6.

fitting value  $\psi_{30\text{keV/u}}/\psi_{135\text{keV/u}} = 0.70 \pm 0.5$  agrees with the prediction within the experimental resolutions. (Here, to estimate  $q_{\parallel}$ , we take the dominant capture channel from the  $^1\Delta$  state to the ground state of helium which gives  $Q \sim 1.1 a.u.$ )

The above analysis shows that the averaged phase shift depends on the projectile velocity. Similar conclusions can be drawn in the regime of  $11.5 \text{ eV} < \text{KER} < 12.5 \text{ eV}$  where the fast repulsive molecular states (the second groups of  $^3\Pi_{\text{III}}, ^1\Sigma^-, ^3\Sigma_{\text{II}}^-, ^1\Delta_{\text{II}}$  etc.) are dominant (see Fig. 2). It is found the phase shifts are  $\Delta'_{30\text{keV/u}} = -0.47 \pm 0.05$  and  $\Delta'_{135\text{keV/u}} = -0.22 \pm 0.07$ , respectively. Within the experimental resolution, their ratio  $\Delta'_{30\text{keV/u}}/\Delta'_{135\text{keV/u}} \approx 2.1 \pm 0.3$  agrees reasonably well with the velocity ratio of 2.1.

We note here that the values of  $\Delta'$  for the second group are different from the those of the first group which reflects the fact that scattering phases for different electronic states are different.

Meanwhile, Figure 3 shows the molecular orientation distributions ( $d\sigma/d\cos\theta = d\sigma/d\Omega$ ) for the KER regime in between 6.0 and 6.5 eV, i.e. from the slow dissociative states of  $^1\Sigma^+$ ,  $^1\Pi$  and  $^3\Pi$ , for collision energies of 30 keV/u and 135 keV/u, respectively. As discussed previously, in slow dissociating processes, the measured molecular orientation does not necessarily reflect the orientation at the instance of the collision. As a result, the orientation distribution turns out to be nearly isotropic for both collision energies.



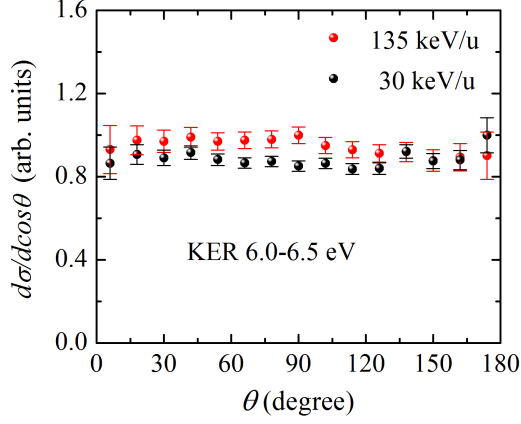


FIG. 3. (Color online) Molecular orientation ( $\theta_R$ ) dependent cross sections of  $6.0 \text{ eV} < \text{KER} < 6.5 \text{ eV}$  for the collision energies of 30 and 135 keV/u from top to bottom.

One should stress that Eqs. (4)-(5), which predict an asymmetry with respect to the angle  $\theta_R = 90^\circ$  due to the presence of the shift  $\Delta$ , refer to fully differential cross sections whereas formula (6) fits the cross section averaged over the momentum transfer. It is, therefore, quite remarkable that even after this averaging the asymmetry is still present. This points to a potentially high sensitivity of the proposed approach to measuring phases. One can therefore expect that in a kinematically complete experiment, for instance on  $\text{CO}^{n+} + \text{He}$  collisions, for which the transverse momentum can be precisely measured, the asymmetry should be even more pronounced. In that case even more detailed information about the scattering phase could be obtained.

The phase shift  $\Delta$  entering Eqs.(4)-(5) depends on the projectile velocity and the transverse momentum transfer which is directly related to the projectile scattering angle  $\theta_p$ . Therefore, by performing measurements at different  $\theta_p$  some control of  $\Delta$  can be achieved by selecting events leading to specific  $\theta_p$  in the data analysis.

$\Delta = \alpha_2 - \alpha_1$  is the relative phase and, as such, in general it does not enable one to identify  $\alpha_1$  or  $\alpha_2$ . However, this restriction can be removed if one chooses the target as a very asymmetric molecule (for instance, HCl or HBr), and properly fits the collision velocity such that one of the  $\alpha$ -s can be made very small.

In conclusion, by exploring double-electron capture in collisions of alpha particles with CO molecules and regarding the atomic cores of the molecule as two “slits”, we have shown that atomic “double-slit” interference can be exploited in order to experimentally extract information about phases of the projectile scattering on the cores of the atoms constituting the molecule. The relative phase can be directly controlled in experiment via modulation of the projectile impact energy.

We expect that similar methods can be also applied to processes like e.g. ionization in order to obtain the information about the corresponding quantum phases. Such a phase con-

trol of collision processes not only can provide the most stringent test for theory but may also open the prelude to new areas of atomic collision studies.

Besides, the ideas expressed here could also be applied to study the nuclear force (which are in some cases not yet very well understood): for instance, by considering neutron scattering on the nuclei of a hetero-DM one can extract information about the corresponding phases of the neutron-nucleus interaction.

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